Ozonesonde Quality Assurance: The JOSIE-SHADOZ (2017) Experience

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Abstract. The ozonesonde is a small balloon-borne instrument that is attached to a standard radiosonde to measure profiles of ozone from the surface to 35 km with ~100-m vertical resolution. Ozonesonde data constitute a mainstay of satellite calibration and are used for climatologies and analysis of trends, especially in the lower stratosphere where satellites are most uncertain. The electrochemical-concentration cell (ECC) ozonesonde has been deployed at ~100 stations worldwide since the 1960s, with changes over time in manufacture and procedures, including details of the cell chemical solution and data processing. As a consequence, there are biases among different stations and discontinuities in profile time-series from individual site records. For 22 years the Jülich [Germany] Ozone Sonde Intercomparison Experiment (JOSIE) has periodically tested ozonesondes in a simulation chamber designated the World Calibration Centre for Ozonesondes (WCCOS) by WMO. In October-November 2017 a JOSIE campaign evaluated the sondes and procedures used in SHADOZ (Southern Hemisphere Additional Ozonesondes), a 14-station sonde network operating in the tropics and subtropics. A distinctive feature of the 2017 JOSIE was that the tests were conducted by operators from eight SHADOZ stations. Experimental protocols for the SHADOZ sonde configurations, which represent most of those in use today, are described, along with preliminary results. SHADOZ stations that follow WMO-recommended protocols record total ozone within 3% of the JOSIE reference instrument. These results and prior JOSIEs demonstrate that regular testing is essential to maintain best practices in ozonesonde operations and to ensure high-quality data for the satellite and ozone assessment communities.

Capsule: Data from ozonesondes form a backbone of satellite algorithms and monitoring stratospheric ozone recovery. The ozonesonde community regularly evaluates sonde procedures and instrumentation, as in this experiment featuring operators from the tropical SHADOZ network.

JOSIE History and Background

The periodic ozone assessments sponsored by WMO/UNEP (1991; 1995; 2011; 2015) and related studies have long recognized the role of ozonesondes in the suite of global observations because sondes are the only technique practical for in-situ monitoring of profiles. The sonde instrument is easy to deploy in remote locations and is relatively inexpensive. Sondes operate in both troposphere and stratosphere (Sidebar 1) and in clouds, precipitation and periods of darkness. Most important, as they ascend, ozonesondes measure ozone with an effective resolution of 100-150 m, far better than satellites. Indeed, sondes, like the ground-based networks of lidar, Dobson and other spectrometers, constitute an essential component of satellite calibration and cross-calibration (Fishman et al., 2008; Hubert et al., 2016; Steinbrecht et al., 2017; Tarasick et al., 2018). The vertical structure of ozone as measured at a typical tropical station appears in Sidebar 1, along with background on ozone in the atmosphere. Although dozens of stations began launching ozonesondes in the 1970s and 1980s, the concepts of standardizing and testing instruments in a coordinated network, did not evolve until the 1990s (Mohnen, 1996; Melamed et al., 2015). This was the period when both the Jülich Ozone Sonde Intercomparison Experiment (JOSIE) and Southern Hemisphere Additional Ozonesondes (SHADOZ) project began.

[Insert Sidebar 1 Here]

Over 50 years of ozonesonde data-taking, there have been several instrument designs. Furthermore, as instruments have changed and preparation and data-processing techniques have evolved over time, time series of data from individual stations often display discontinuities and gaps that lead to inhomogeneous data records. Thus, the reliability of ozonesonde trends was questioned in some of the earlier ozone assessments (WMO/UNEP 1991; 1995; SPARC/IOC/GAW, 1998) (See Acronym List).

Two approaches have been used to address these deficiencies. First, evaluations of ozonesonde types in a controlled laboratory environment were undertaken in the 1990s, a process that continues periodically to this day. Second, in a similar manner, by testing different sonde preparation methods and protocols for data recording and processing, a set of standard operating procedures (SOP; Smit et al., 2014) was developed through consensus with the ozonesonde research community. Finally, there are recommended methods for reprocessing long-term records compromised by inhomogeneities (Smit et al., 2012, Deshler et al., 2017).

The need to have recommended instruments and procedures for emerging WMO/GAW stations in the 1990s provided a framework for the first intercalibration and intercomparisons of existing ozonesonde types. In order to assess the performance of the various ozonesonde instrument types used within GAW, the environmental simulation chamber (ESC) at the Forschungszentrum Jülich (FZJ, Germany) was established as the World Calibration Centre for Ozone Sondes (WCCOS) in 1996. The chamber enables control of pressure, temperature, and ozone concentration as it simulates flight conditions of ozone soundings up to an altitude of 35 km (Smit et al., 2000). This controlled environment and comparison of the ozonesonde profiles with an accurate UV-photometer as a reference (Proffitt and McLaughlin, 1983) are essential requirements for addressing instrument issues that arise from field and laboratory operations.

The initial JOSIE, performed in 1996 (Smit and Kley, 1998), was the first GAW activity directed toward implementing a global quality assurance plan for ozonesondes in routine use. By now, JOSIE experiments have provided over twenty years of ozonesonde data quality assurance to the larger atmospheric research and remote sensing communities. JOSIE-1996 was attended by eight laboratories from seven countries representing the major types of ozonesondes:

Electrochemical Concentration Cell (ECC) sondes of two manufacturers, the Brewer/Mast sonde (BM-original), the Indian sonde (a modified BM-type), and the Japanese Meisei sonde (KC79). JOSIE-1996 revealed important information not only about ozonesonde performance but also the influence of operating procedures for sonde preparation and data correction that often varied among the participating laboratories. The succession of JOSIE campaigns (**Table 1**) has shown that there is an on-going need to evaluate ozonesondes because the instruments, preparation procedures, and/or the sensing solutions are modified, often inadvertently, over time. Routine testing of newly manufactured ozonesondes on a regular basis coupled with better standardization of operating procedures help ensure more confidence in the data itself as well as trends calculated from the data.

The overall objective of WCCOS and the JOSIE series of experiments has been the establishment of a facility for ozonesonde quality assurance (QA) that can be used by sonde manufacturers and the research community. Instrumental performance of sondes from different manufacturers is tested through comparison of profiling capabilities with a standard ozone profile that simulates a typical ascent in polar, mid-latitude or tropical conditions. Regular evaluation of procedures and methods at long-term ozone sounding stations with a single ozone reference instrument ensures the traceability and consistency of the records.

Over time, the SOP have been established and updated as needed. The first major SOP documentation appeared as a WMO/GAW Report (#201; See Smit and ASOPOS, 2014) with major contributions from prior reports and Smit et al. (2007). GAW 201 was also based on field tests of the major sonde types used in the JOSIEs up through 2009. A gondola of 18 instruments was flown along with same UV-photometer used in JOSIE-2000 as reported in Deshler et al. (2008).

SHADOZ and Unresolved Sonde Issues

The SHADOZ network began in 1998 as an international partnership to enhance the number of tropical ozone soundings from operational stations (Thompson et al., 2003a,b; 2004; 2007; 2011). SHADOZ uses ECC ozonesondes that, over time, have been coupled with a variety of radiosondes (**Table 2**). A history of ozonesonde-radiosonde pairings used at SHADOZ sites appear in archival papers (Thompson et al., 2003a,b; Thompson et al., 2007; Witte et al., 2017). At the time SHADOZ began, all known operational stations were in the southern hemisphere, but gradually northern hemisphere stations joined: Kuala Lumpur, Paramaribo, Costa Rica; Hanoi, and Hilo. The 14 long-term stations, defined as operating at least a decade during SHADOZ, appear in **Fig. 1.** More than 7000 sets of ozone and pressure-temperature-humidity profiles from SHADOZ are available at the website: https://tropo.gsfc.nasa.gov/shadoz.

Periodic evaluations of SHADOZ data have examined three parameters. First, total column ozone (TCO) from the sonde, with an appropriate extrapolation above balloon burst, e.g., McPeters and Labow, (2012), is compared to TCO from co-located ground-based instruments (Brewer, Dobson, SAOZ) and satellite overpasses. Second, stratospheric profiles are compared to satellite overpass ozone profiles from instruments like SAGE II (to 2005), SBUV (entire record, 1998-2016) or Aura's MLS (2005-). Third, for the tropical stations (generally within 18° latitude of the equator), stratospheric column ozone and profiles are compared. The tropical TCO is typically constant to within 3-5 DU (Dobson Units), so measurement biases from station to station can be identified (Thompson et al., 2017).

The first three years of SHADOZ TCO compared to the EP/TOMS satellite TCO disagreed by ~8% on average, with a number of stations displaying a discrepancy of greater than 10%; the sonde TCO was usually lower than the satellite (or ground-based instrument). After the JOSIE-2000 campaign (Smit et al., 2007), in which the instruments and techniques used at all the

SHADOZ stations were tested, several stations changed their sensing solution type (SST), resulting in reduced offsets (Thompson et al., 2007). Further changes in sonde preparation procedures and subsequent reprocessing of the data, both in accordance with WMO/SPARC/IOC/NDACC guidelines (Smit and O3S-DQA, 2012; Smit and ASOPOS, 2014), brought TCO for 12 of 14 stations to within 2% of TCO from three BUV-type satellites (EP/TOMS, OMI and OMPS) operating over the 1998-2016 period (Thompson et al., 2017); the remaining two stations show TCO data averaging within 5% of the satellite TCO. These improvements derive from the application of "transfer functions" that relate a profile from each instrument–SST combination to data from the standard reference. Each profile in a time-series is examined for possible correction (Witte et al., 2017; 2018).

Although the reprocessing of prior SHADOZ data has greatly reduced systematic variations in the record, JOSIE-SHADOZ was designed to address several outstanding issues. First, transfer functions determined by Deshler et al. (2017) are used to homogenize SHADOZ readings that are taken with different SST and/or instruments. This includes the 1%, KI, 0.1% buffer SST used at stations supported by NOAA since the mid-2000s (Sterling et al., 2018). Second, a few stations in SHADOZ changed SST unintentionally and introduced discontinuities in station time-series (Thompson et al., 2017; Witte et al., 2017; 2018). Finally, several stations employing a given sonde type show sharp discontinuities after 2014 that appear to originate with changes in manufacture (Sterling et al., 2018; Thompson et al., 2017).

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JOSIE-SHADOZ-2017 Goals

Similar to prior JOSIE campaigns, the major objectives of JOSIE-SHADOZ are:

1. Evaluate ozonesonde instrument performance, specifically the pump and sensor as delivered

by the ECC-sonde manufacturer. Most of the SHADOZ stations operate with WMO-recommended solutions and preparation and calibration procedures that allow the experimenters to update typical performance of the instruments relative to the Ozone Photometer (OPM) reference instrument (Proffitt and McLaughlin, 1983).

2. Evaluate current preparation and operating procedures of each SHADOZ station. Unlike prior JOSIE experiments, in 2017 personnel representing the practices of all currently operating SHADOZ stations participated (**Tables 2 and 3; see Sidebar 2)**. In most cases the operators supplied solutions as prepared at their home institution. In the first part of the JOSIE-2017, the operators followed their standard practice for pre-conditioning sondes and for "day of flight" prior to simulation in the ESC. The goal was to understand the existing ozone profiles archived in SHADOZ by reproducing current practices, techniques, and solutions at each participating station as closely as possible.
3. Evaluate the current WMO recommended SOP. Specific instrumental aspects examined in these tests were details of pre-conditioning, background current, response time, pump flow efficiency, and SST. In addition to two WMO-recommended SST, two alternatives, one of which is employed at several SHADOZ stations, were included in the tests.

The Ozonesonde Design

The electrochemical concentration cell (ECC) ozonesonde uses a chemical reaction measured inside a pair of cells that is displayed schematically in **Fig. 3a.** As the sonde rises in the atmosphere (and during the laboratory calibration phase), air is pulled through the intake tube (left in **Fig. 3a)** and pushed into the cathode cell by means of a small pump. The pump maintains positive pressure as the air is sampled; the flow rate is measured during pre-flight calibration. The second cell (anode) is filled with a saturated version of the cathode solution and is located adjacent to the cathode, with an ion bridge separating the two cells. The reacting chemical, oxidized by the ozone molecule, is dissolved potassium iodide (KI). The sensing solution is maintained at a neutral pH with the addition of the paired phosphates ($NaH_2PO_4 \bullet H_2O$/ $Na_2HPO_4 \bullet 12H_2O$). The ozone partial pressure is calculated by the following equation (taken from Witte et al. 2018),

$$P_{O3} = 4.307 \times 10^{-2} \frac{(I_M - I_B) T_P}{\Psi_P \Phi_P \eta_C}$$

where

P_{O3} = Ozone partial pressure, mPa

I_M = Cell current, μA

I_B = Cell background current, μA

T_P = Ozonesonde pump temperature, K

Φ_P = Pump flow-rate, ml/s

Ψ_P = Pump flow efficiency, unitless

η_C = Conversion efficiency which is generally assumed to be 1.

The pump flow efficiencies, Ψ_P, take into account the buffering of the solution, depending on the solution recipe, and mechanical degradation of the pump at low pressures (< 100 hPa). The volume mixing ratio is computed from the ratio of the ozone partial pressure (P_{O3}) to the ambient pressure determined from the radiosonde attached to the ozonesonde container as the two instruments ascend into the stratosphere (**Fig. 3b**). The typical ascent rate is 5 m/s.

From the large body of SHADOZ data as well as instruments in the field and prior lab intercomparisons it is known that the two major sources of systematic error are the manufacture of the instrument and the composition of the KI and/or buffers in the SST (Smit et al., 2007).

Random sources of error include operator handling and changing conditions in the station calibration unit. Calibration practices and the method of data-processing can also lead to systematic differences among station profiles (Johnson et al., 2002; Deshler et al., 2008; 2017). In JOSIE-SHADOZ two types of protocols investigated these issues. The first five of ten tests in each session were carried out with the operators using their own solutions and preparation technique. We refer to this as SHADOZ SOP (Standard Operating Procedure). In the second set of tests, uniform calibration and preparation procedures were followed using JOSIE-prepared solutions, hereafter referred to as the JOSIE SOP. Unified data collection by the Data Acquisition System (DAS) eliminates variations due to operator data-processing.

General Operations during JOSIE-SHADOZ (2017). The JOSIE-SHADOZ 2017 campaign took place at the World Calibration Centre for Ozone Sondes (WCCOS) at the Research Center Jülich (FZJ) in the Institute of Energy and Climate Research: Troposphere (IEK-8), Jülich, Germany. Ozonesonde pre-conditioning test units and the ECC instruments were provided by FZJ from a pool of loaned supplies. Participants were split into two groups (**Table 3**), each of four teams operating ozonesondes of the type used in SHADOZ (**Table 2**). Each group participated in a 12-day intercomparison campaign. Session No. 1 took place from 9 to 20 October 2017; Session No. 2 took place from 23 October through 3 November 2017. Each session consisted of ten simulation experiments with all four participant sondes being “flown” simultaneously in the chamber (see **Sidebar 3**) to an effective altitude of ~35 km. The overall protocol for each campaign was similar but the second session tested two “JOSIE SSTs” (**Table 4**). During the SHADOZ SOP (first five simulations) participants used their own zero-air filter, solutions, and preparation procedures. During the JOSIE SOPs the lab provided a single source of high-quality zero-air, a common SST, and common operating procedures that all teams followed. Data were collected by the DAS of the WCCOS test chamber.

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Because JOSIE-SHADOZ 2017 was focused on questions about SHADOZ operations, all the chamber runs simulated tropical sounding conditions (**Fig. 4)**. The test profiles described in **Fig. 4** and **Table 4** represent three typical tropical profiles, one that is unpolluted throughout the troposphere with very low ozone near the tropopause and two with higher levels of ozone in the free troposphere and near the tropopause.

Four SST recipes were tested. All sonde data were processed by using a constant background current correction. Total ozone column normalization was not applied. The solutions, with references, follow:

1. SHADOZ 1.0. The WMO-recommended SOP (Smit et al., 2012) for use with the Science Pump (SPC) instrument and is referred to as SST 1.0%-Full Buffer:

   Cathode: 1% KI + Full-Buffer & KBr as described by Komhyr (1986)

   Anode: Cathode solution with saturated KI

   Pump flow efficiency factors (PEF): Komhyr (1986)

2. SHADOZ 0.5. The WMO-recommended SOP (Smit et al., 2012) for use with the ENSCI instrument is referred to as SST 0.5%-Half Buffer:

   Cathode: 0.5% KI + Half of the Buffer & KBr as described by Komhyr et al. (1995)

   Anode: Cathode solution with saturated KI

   PEF: Komhyr et al. (1995)

3. JOSIE 1.0.1. Solution developed by NOAA for use with ENSCI sondes that has been employed at Fiji, Samoa, Costa Rica, and Hilo stations since the late 2000’s. The formulation is SST 1.0%-1/10th Buffer:

Cathode: 1% KI+ $1/10^{th}$ Buffer, KBr as described by Komhyr (1986)

Anode: Cathode solution with saturated KI

PEF: New constants derived from recent pumpflow measurements made by Nakano (2017, private communication).

4. JOSIE 2.0.1. This variation on JOSIE 1.0.1 was used to test if ozone response in the tropopause and stratosphere regions is improved by doubling the KI concentration:

Cathode: 2% KI + $1/10^{th}$ Buffer, KBr as described by Komhyr (1986)

Anode: Cathode solution with saturated KI

PEF: New constants derived from recent pumpflow measurements made by Nakano (2017, private communication).

Preliminary Results

Preliminary data are used to answer three questions. (1) What is the accuracy of ozone readings throughout the profile for each sonde-SST combination tested in the ESC? This is answered by comparing both the ozone partial pressure profiles measured by the sonde with the OPM and column-integrated ozone from the sondes with the OPM. For the latter, TCO and segments for troposphere, stratosphere and the tropopause transition layer (TTL) in between the stratosphere and troposphere are computed. (2) How do profiles and column segments from sondes prepared with the SHADOZ SOP compare to those prepared with the JOSIE SOP? (3) What differences are observed when the same instrument type is prepared with different SST or when different instruments use the same SST? Differences are expected based on prior JOSIE results and field tests.

SHADOZ SOP. **Fig. 5** displays raw data from eight SHADOZ participants. The OPM measurements are represented by the black dashed lines: **Fig. 5a** shows the data for a simulation in Session 1 (No. 171) and **Fig. 5b** for a simulation in Session 2 (No. 182). The fundamental unit in the tests is lapsed time; quoted "altitudes" are approximate. There is some arbitrariness in designating the TTL, with lower-mid-troposphere below and mid to upper stratosphere above. We adopt a TTL at 2200-3800 s (~12-18 km) when analyzing the test results. In this region the signal-to-noise ratio is low, and therefore the uncertainty, is highest (Witte et al., 2017).

In **Fig. 5a** the ozone partial pressures are very small throughout the "troposphere" and up to ~3500 s or ~17.5 km. This profile simulates a near-zero-ozone tropopause, mimicking western Pacific profiles (Kley et al., 1996; Thompson et al., 2012; Rex et al., 2014; Newton et al., 2016), where SNR in ozone readings is often low. In **Fig. 5b** ozone partial pressure throughout the tropospheric profile is higher, representing stations influenced by biomass burning pollution in the lower-mid troposphere (Thompson et al., 1996; Jensen et al., 2012). The ozone transition near the tropopause and in the lower stratosphere in Simulation No. 182 (**Fig. 5b)** lacks the sharp gradient intentionally generated in **Fig. 5a.** The pattern in **Fig. 5b** resembles that of SHADOZ stations that exhibit gradual ozone transitions in the TTL, e.g., Ascension, Natal and Nairobi. Their upper tropospheric and TTL cross-sections and their contributions to the zonal wave-one in tropical ozone are summarized in Thompson et al. (2003b; 2011; 2017).

The OPM TCO in **Fig. 5a** is 282 DU. The TCO from the four participants in Session 1 are all higher than the OPM by 3-26 DU (up to 9%). The OPM TCO in **Fig. 5b** is 334 DU. The TCO from the four participants in Session 2 are all equal to or higher than the OPM, with the largest offset 23 DU (7%) higher. Columns 2 and 3 in **Table 5** list the corresponding TCO fractions for all 8 participants relative to the OPM.

The means of five simulations for all eight participants, expressed as absolute and percentage differences from the OPM and based on their SHADOZ SOP are displayed in **Fig. 6.**

The shapes of the mean profiles are broadly similar with the sonde partial pressures (relative to the OPM, **Fig. 6a)** overlapping throughout the troposphere and TTL (to 3500s). In the stratosphere (above 4000 s, ~20 km) differences are much larger. The fractional differences are smaller in the stratosphere (**Fig. 6b)**, however, because the ozone partial pressure peaks at over 20 mPa (**Fig. 5).** The relative differences with the OPM are largely within ± 10% of the OPM (zero-line in **Fig. 6b**) throughout the lower to mid-troposphere (0-2000 s, up to 10-12 km). Around 2000 s, there is an inflection, with the offsets all turning more negative. The largest relative differences occur within the upper troposphere (UT) and TTL (equivalent to 2500-3500 s, 13-18 km), exceeding 5% on average for all the stations. For participant nos. 4 and 5 the mean relative differences exceed -20%. Witte et al. (2018) noted that SHADOZ ozone values are most uncertain in the narrow region between 15 and 17 km (~3000-4300 s). However, the large offsets recorded in **Fig. 6b** originate from four JOSIE tests conducted with TTL ozone equivalent to 2 DU (e.g. Simulation 171, **Fig. 5a**); a value that applies to only ~ 5% of tropical SHADOZ readings. Realistically, Fig. 8b in Thompson et al. (2017), based on > 6000 profiles, shows that the actual TTL ozone for 12 of 14 SHADOZ stations is 8.0±1.5 DU. By 3000 s (~15 km) the relative differences of all SHADOZ profiles with respect to the OPM start to increase. All SHADOZ profiles show excellent agreement with OPM to within ± 5% at 20-25 km (critical ozone maximum). By 5000 s (~ 25 km) most SHADOZ profiles exceed OPM ozone and are well-aligned with one another. The range of mean deviations in the region corresponding to 20-28 km is within 10%. This tighter clustering implies good measurement precision. By ~5500 s (27.5 km) all the SHADOZ readings are higher than the OPM. Above 30 km the agreement breaks down and there is a downturn in ozone readings relative to the OPM for most stations. Exceptions are participant No. 1 and 7 that display +10% and 4% deviations, respectively (**Fig. 6b**). The negative relative differences are not surprising. Witte et al. (2017) showed that even reprocessed SHADOZ ozonesonde data above ~30 km are highly variable and not as reliable.

How do column amounts for the SHADOZ participants compare on average to OPM ozone? Answers appear in **Table 5.** For the five SHADOZ simulations all of the participants record, on average, slightly more ozone than the OPM, with ratios from 1.017-1.040 (1.7% to 4.0% more O_3). This result seems to validate the quality assurance practices of the SHADOZ stations, with 7 of 8 participants following the WMO-recommended instrument SST combinations and SOP (Smit et al., 2007; 2012). The segment column comparisons (columns 0-15 km, 12-18 km, 15 km-end in **Table 5)** demonstrate that the good agreement between sondes and the OPM is dominated by the ozone column from 15 km-end, i.e., the stratospheric portion of the profile. Because the WMO recommendations are largely based on JOSIE-2000, several follow-on lab tests and the BESOS conducted in 2004, it can be inferred that the WMO recommendations (Smit et al., 2012) are still valid. Agreement in the TTL (12-18 km column) averages < 0.95 for half of the groups (**Table 5)**. Because the OPM recorded only 5 DU on average in this region, the larger offsets do not detract from the good agreement overall.

JOSIE SOP. The sonde partial pressure offsets from the OPM and relative differences for the eight participants using the JOSIE 1.0.1 SST and preparation protocols appear in **Fig. 7a** and **Fig. 7b**, respectively. When these results are compared to those with the SHADOZ SOP (**Fig. 6)** two differences are observed. First, the divergence among stations is less with the more uniform specifications of the JOSIE SOP, especially in the mid-troposphere through the TTL. This is not surprising because the use of a single SST and SOP is expected to minimize variations due to SST. The JOSIE SOP uses solutions with less buffer by a factor of 2 or 10. Thus, due to the lower buffer the sonde responses show less hysteresis effect in the region with relatively fast ozone changes, resulting in increased SNR. This is particularly true in the TTL at the tropopause and just above, corresponding to the 2500 to 3500 s region in **Fig. 6b** and **Fig. 7b**.

The second difference is that ozone readings throughout the profile are lower relative to the OPM with the JOSIE SOP than the SHADOZ SOP, particularly in the troposphere (**Fig. 7a** below 4000 s) and even more so in the stratosphere, where the offsets are -1 to -2 mPa ozone. The result is a mean sonde TCO offset with the JOSIE SOP relative to the OPM of 0.97 (first two entries in column three of **Table 6)** compared to a mean 1.03 TCO offset with the SHADOZ SOP. Background cell currents and response times improved significantly during the JOSIE SOP in both sessions when a shared zero-air system was used.

SHADOZ-JOSIE Comparisons. **Fig. 8a** displays the average differences between the SHADOZ and JOSIE SOP profiles for Session 1. For each participant in Session 1, five simulations were made totaling 20 profiles of each SOP, both using the same SST. Up to 10 km the SHADOZ SOP resulted in relatively higher ozone readings; toward the TTL the JOSIE SOP resulted in higher ozone readings. The stratospheric differences, however, show the JOSIE SOP averages 3% lower TCO than the OPM while the SHADOZ SOP averages 3% higher TCO than the OPM (and stratospheric segment, **Table 6**). Note that the near-zero simulated ozone represents a small fraction of what is observed in SHADOZ records; thus, the large uncertainties seen in **Fig. 8a** represent the extrema of the data set.

In Session 2, to compensate for the reduced sensitivity of the 1.0%, 1/10th Buffer SST (JOSIE 1.0.1), solutions with the JOSIE SOP were prepared with twice as much KI but the same low buffer, the so-called JOSIE 2.0.1. JOSIE 1.0.1 comparisons were all made with ENSCI, whereas the JOSIE 2.0.1 referred to a combination of SPC and ENSCI. Mean profile comparisons with the different SSTs are summarized in **Fig. 8b**. The differences are not statistically significant throughout the troposphere or TTL but the JOSIE 2.0.1 profile mean is closer to the OPM in the upper stratosphere (above 5000 s). In Session 2, the ratio of sonde to OPM partial column ozone above 20 km for JOSIE 1.0.1 was 0.95, while for JOSIE 2.0.1 it was

0.97. Sondes filled with both SST show sondes measure less ozone than the OPM in the stratosphere and are highly variable above 30 km, consistent with **Fig. 7** and Witte et al. (2018) findings.

Previous JOSIE campaigns and various field tests (especially the BESOS in 2004) noted that throughout the ozone profile when the same SST is used, the ENSCI instrument tends to measure more ozone than the SPC instrument. Of the 14 SHADOZ stations, 11 use the ENSCI instrument and three use the SPC type (Thompson et al., 2017; Witte et al., 2017; 2018). **Fig. 8c,** based on the combined session simulations (JOSIE 1.0.1), shows that, also for the less buffered solutions, the ENSCI instrument measures slightly higher ozone than the SPC with the greatest discrepancies in the troposphere, consistent with previous JOSIE studies.

Conclusions

1. All 8 stations participating in JOSIE-SHADOZ-2017 measured ozone that agreed well with the OPM.
2. The slight ENSCI – SPC ozone bias (ENSCI reads higher) previously observed (Smit et al., 2007; 2012) remained in JOSIE-SHADOZ 2017.
3. JOSIE-2017 affirms the very high quality of the SHADOZ methods that use SOP and SST-instrument combinations based on earlier JOSIE campaigns and field tests as summarized in Smit et al. (2007; 2012). This is independent confirmation of the accuracy of the large SHADOZ dataset that up to now has only been compared to data from satellite and ground-based instruments (Thompson et al., 2017; Witte et al., 2017). The ozonesonde community goals of “5% accuracy and precision in TCO” has been met by SHADOZ operators engaging in collaborative ozonesonde “expert” activities since 2000. Except for the TTL, most instrument-SST combinations tested in JOSIE with

SHADOZ SOP agreed within 3% of OPM in total column amount (sonde higher) and 5-10% throughout the ozone profile. The often large TTL ozone underestimate (>30% relative to OPM in some tests) contributes only 2-3% of the total ozone column.

4. JOSIE tested solutions with a reduced buffer SST, of the type used at four SHADOZ stations. As expected, agreement of sonde ozone data with the OPM in the TTL regions was improved. However, sensitivity to stratospheric ozone is reduced, so TCO from these tests averaged 3% lower than the OPM. The low-bias is reduced when the KI is doubled (JOSIE2.0.1). However, the divergence of profiles with the different SST is so small (~5%) that further analysis, such as taking into account individual sonde responses, is required.
5. JOSIE SOP:
    - Lower, uniform, and better reproducible background cell currents are achieved using a high quality no-ozone filter source or purified air.
    - The hysteresis effect (‘memory’ effect due to the buffering of the solution) is minimized which may improve the response of the sonde, particularly in the TTL where sharp ozone gradients are measured.

Because SHADOZ represents virtually all current ECC sonde practices used by the global ozone community, these findings and any SOP recommendations that ozonesonde “experts” consider in light of JOSIE-2017 should be universally valid for ECC instruments. Establishing SOP guidelines and standardization of ground equipment is essential to achieving an uncertainty less than 5% between surface and 30 km altitude. The JOSIE-SHADOZ 2017 experience highlights the necessity of having a continuous reference calibration facility (WCCOS) operating over the past 25 years. The capacity building exercise has empowered participants to continue working towards ensuring high quality standard in sonde data-taking. With well-trained and motivated operators, SOPs based on best practices, and experiments such as JOSIE-SHADOZ, our aim of an uncertainty less than 5% can be achieved.

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ACRONYMS

ASOPOS = Assessment of SOP for Ozone Sondes

DAS = Data Acquisition System

ECC = Electrochemical Concentration Cell

ENSCI = Environmental Science Corp.

ESC = Environmental Simulation Chamber

FZJ = Forschungscentrum - Jülich

GAW = Global Atmospheric Watch

GSFC = Goddard Space Flight Center

IOC = International Ozonesonde Commission

JAMSTEC = Japan Agency for Marine-Earth Science and Technology

JOSIE = Jülich Ozonesonde Intercomparison Experiment

KNMI = Koninklijk Nederlands Meteorologisch Instituut

MLS = Microwave Limb Sounder

NDACC = Network for the Detection of Atmospheric Composition Change

OPM = Ozone Photometer

OPS = Ozone Profile Simulator

PEF = Pump Efficiency Factor

QA = Quality Assurance

SBUV = Solar Backscatter Ultraviolet

SHADOZ = Southern Hemisphere Additional Ozonesondes

SNR = Signal Noise Ratio

SOP = Standard Operating Procedures

SPARC = Stratospheric Processes And their Role in Climate

SPC = Science Pump Corporation

SST = Sensing Solution Type

TCO = Total Column Ozone

TTL= Tropical Tropopause Layer (or Tropopause Transition Layer)

UNEP = United Nations Environmental Programme

WMO = World Meteorological Organization

WCCOS = World Calibration Centre for Ozonesondes

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Sidebar 1: Ozone in the Earth's Atmosphere

The ozone molecule (O_3) plays several important roles in the earth's atmosphere. Its absorption of radiation warms the stratosphere, leading to the temperature inversion between troposphere and stratosphere (**Fig. SB1**). The inversion is typically referred to as the tropopause but we use the term "tropopause transition layer" to signify that the tropopause is a region (~130-70 hPa) in which a number of physical properties gradually change. Eighty-ninety percent of the ozone molecules reside in the stratosphere so harmful uv radiation is blocked from reaching the earth's surface. In the free troposphere, ozone acts as a greenhouse gas and is estimated to be responsible for ¼ to 1/3 of earth's warming over the past 200 years. Tropospheric ozone is also a source of the OH free radical, the primary oxidant in the atmosphere, responsible for reacting with hundreds of species (Thompson, 1992). Ozone at the surface is considered a pollutant, harmful to human and plant health when it exceeds 3 mPa (**Fig. SB1**).

Sidebar 2: Capacity building during JOSIE-SHADOZ

A unique feature of JOSIE-SHADOZ was that the ozonesondes were prepared by operators from organizations representing eight SHADOZ sites (see **Fig. 2** showing group photos taken during both sessions in front of the WCCOS chamber). Capacity-building activities during both sessions included lectures on sonde quality-assurance, the importance of metadata reporting, troubleshooting, and training with coaches from sponsoring organizations: NASA/GSFC; NOAA/GMD; KNMI (Netherlands); KMI (Belgium); Meteoswiss, Environment – Climate Change Canada; the Finnish Meteorological Institute. Financial support for the tropical operators came from the UNEP-sponsored Vienna Convention Trust Fund, administered by WMO. Operators are essential contributors to ozonesonde quality assurance by providing detailed metadata information on each sonde launch and maintaining uniformity in their preparation and launch procedures. Bringing together SHADOZ operators for training and knowledge sharing helps to ensure that best practices are applied to operations in a consistent manner across the SHADOZ network.

Sidebar 3: Design of the ESC, Reference Instrument, Data System.

The WCCOS, the only one of its kind, was established in the mid-1990s at FZ-Jülich to test, calibrate and compare different types of balloon borne ozonesondes that are used to measure the distribution of ozone in the troposphere and lower/middle stratosphere. The facility is described in more detail in Smit et al. (2000): http://www.fz-juelich.de/iek/iek-8/EN/Expertise/Infrastructure/ESF/ESF_node.html.

The setup of the simulation facility (Fig. **SB2a),** consists of four major components:

1. <u>Environmental Simulation Chamber</u>. The ESC chamber is a temperature-controlled vacuum chamber with a test room volume of about 500 liter (80 x 80 x 80 cm). Within the ESC the pressure and temperature can be dynamically regulated, with pressures between 5 and 1000 hPa and temperatures between 200 and 300 K, with a maximum rate of ±2K/min. Iso-thermically operated, the temperature variations of the air as well as the wall inside the test room can be maintained within ± 0.2 K. For more details see Smit et al. (2000).
2. <u>Ozone Photometer (OPM), Ozone reference.</u> The OPM is a fast response dual-beam UV-absorption photometer, originally developed by Proffitt and McLaughlin (1983) for use on stratospheric balloons. The instrument was flown during Balloon Ozone Intercomparison (BOIC) missions in 1983/1984 (Hilsenrath et al., 1986); it was used in the Balloon Experiment on Standards for Ozone Sondes (BESOS) field campaign in Wyoming, in 2004 (Deshler et al., 2008). The OPM is an absolute measuring device with a 1-s response time at a sampling volume flow rate of about 8 l/min. The overall accuracy of ozone measurements

made by the OPM is better than ±2% for simulated altitudes up to 25 km (pressures down to 25 hPa) and ±3.5 % at 30-35 km altitude (12-5 hPa). The instrument resides in a separate vacuum vessel which is connected to the ESC such that the UV-photometer has the same pressure conditions as inside the test chamber.

3. Ozone profile simulator (OPS). A gas-flow system that controls the ozone concentrations sampled by the instruments in the ESC, with a gas flow rate of 12-15 l/min. The OPS can simulate vertical ozone profiles between the surface and 35 km. The OPS can accommodate up to four ozonesondes, including the OPM (Fig. **SB2b)**. The OPS has an option to specify ozone step functions or zero ozone to investigate the response time and background characteristics of ozonesondes.

4. Data Acquisition System (DAS). The entire simulation process is automated by computer control in order to have reproducible conditions with respect to the simulated pressure, temperature and ozone versus time, and for recording and storing the large variety of parameters measured during the simulation process. A special electronic interface (JOSIE/ ECC-interface) couples the ECC sonde to the DAS, transmitting cathode cell current, pump temperature, pump motor current and pump motor voltage (12V). A small variable electrical heater (0-10W) adjusts pump temperatures to values similar to actual flight temperatures.

Table List:

Table 1: JOSIE activities on ozonesonde procedures and related reports.

Campaign	Objective
JOSIE-1996 **GAW Report #130**	• Operating Procedures • Profiling Capabilities • Intercomparison sonde types (ECC, Brewer Mast, Meisei)
JOSIE-1998 **GAW Report #57**	• Manufacturing ECC sondes (SPC, ENSCI)

JOSIE-2000
GAW Report #158
(Smit et al., 2007)
	• Operating Procedures
	• Focus on ECC sonde
	  ○ Different sensing solution types
	  ○ Different manufacturers (SPC, ENSCI)

BESOS-2004
(Deshler et al., 2008)
	• Operating Procedures under flight conditions
	• Focus on ECC sonde
	  ○ Different sensing solution types
	  ○ Different manufacturers (SPC, ENSCI)

ASOPOS 2002-2012 **GAW Report #201**	• Define and establish Standard Operating Procedures for ECC sondes
JOSIE-2009	• Manufacturers (SPC, ENSCI)
JOSIE-2010	• Refurbished sondes
O3S-DQA Guidelines **Report-2012**	• Homogenization and Uncertainties
JOSIE-SHADOZ-2017	• Operating procedures • Tropical simulations • Different sensing solution types • Different manufacturers (SPC, ENSCI)

Table 2: SHADOZ stations operating at least 10 years between 1998 and 2017

Station	Latitude, Longitude	Current ECC Sensor	Current Radiosonde
Pago Pago, Am. Samoa	14.23S, 170.56W	ENSCI	iMet-1
Hilo, Hawaii	19.40N, 155.00W	ENSCI	iMet-1
San Cristobal, Galapagos, Ecuador	0.92S, 89.60W	ENSCI	Vaisala RS92
San Pedro, Costa Rica	9.94N, 84.04W	ENSCI	iMet-1
Paramaribo, Surinam	5.81N, 55.21W	SPC	Vaisala RS92
Ascension Is., U.K	7.98S, 14.42W	ENSCI	iMet-1
Natal, Brazil	5.42S, 35.38W	SPC	Lockheed-Martin-Sippican LMS6
Irene, S. Africa	25.90S, 28.22E	SPC	Vaisala RS92
Nairobi, Kenya	1.27S, 36.80E	ENSCI	Vaisala RS92
La Réunion, France	21.10S, 55.48E	ENSCI	Modem M10
Kuala Lumpur, Malaysia	2.73N, 101.70E	ENSCI	GRAW DFM-09
Hanoi, Vietnam	21.02N, 105.80E	ENSCI	Vaisala RS92
Watukosek-Java, Indonesia	7.57S, 112.65E	ENSCI	---*
Suva, Fiji	18.10S, 178.40E	ENSCI	iMet-1

*Operated Meisei RS II-KC79D radiosonde-ozonesonde system 1992-1999; Vaisala RS80 1998-2013.

Table 3: SHADOZ station operators and instruments tested in JOSIE. Stations 1-4 participated in Session 1 (9-20 October 2017); stations 5-8 participated in Session 2 (23 October – 3 November 2017).

Participant Number	SST	Operator	Affiliation	Station
		Session 1		
1	1.0% Full Buffer	Tshidi Machinini	South African Weather Service	Irene, South Africa
2	1.0% Full Buffer	Francisco R. da Silva	Brazilian Space Agency	Natal, Brazil
3	0.5% Half Buffer	Kennedy Thiong'o	Kenyan Meteorological Department	Nairobi, Kenya
4	0.5% Half Buffer	Ernesto Corrales	University of Costa Rica	San Pedro, Costa Rica
		Session 2		
5	1.0% Full Buffer	George Paiman	Meteorological Service of Suriname	Paramaribo, Surinam
6	0.5% Half Buffer	Zamuna Zainal	Malaysian Meteorological Department	Kuala Lumpur, Malaysia
7	0.5% Half Buffer	Françoise Posny	Université La Réunion, Météo-France, CNRS	La Réunion Is., France
8	0.5% Half Buffer	Nguyen Thi Hoang Anh	Vietnam Meteorological and Hydrological Administration	Hanoi, Vietnam

Table 4: Characteristics of JOSIE-SHADOZ-2017 simulations in the WCCOS chamber with Simulation Numbers listed for the two Sessions. LT=lower troposphere, MT=mid-troposphere, UT=upper troposphere and LS=lower-stratosphere. All profiles simulated with nominal 5 m/s ascent velocity. The tropopause was located at Z=18-20 km with minimum temperature ~-(70-80)$^{\circ}$C. The stratospheric profile was specified to be the same for all simulations.

			Session 1	
Simulation Number	**Troposphere Profile Type**	**Profile Type Index****	**Specifications**	**ECC Procedure**
171	Recent deep convection	1	Extremely low O_3 values nearly uniformly up to tropopause with very steep gradient into LS	Station-supplied SST & procedures
172	Maritime background	2	Low O_3 in LT, moderate O_3 in MT, extremely low O_3 in UT	Station-supplied SST & procedures

173, 174, 175, 176*	Biomass burning	3	Enhanced O_3 in LT, high O_3 in MT, low O_3 in UT	Station-supplied SST & procedures
177, 178, 179, 181	Biomass burning	3	Enhanced O_3 in LT, high O_3 in MT, low O_3 in UT	JOSIE-supplied SST & WMO procedures
180	Maritime background	2	Low O_3 in LT, moderate O_3 in MT, extremely low O_3 in UT	JOSIE-supplied SST & WMO procedures
Session 2				
Simulation Number	**Troposphere Profile Type**	**Profile Type Index****	**Specifications**	**ECC Procedure**
182, 183, 184,186	Biomass burning	3	Enhanced O_3 in LT, high O_3 in MT, low O_3 in UT	Station-supplied SST & procedures
185	Maritime background	2	Low O_3 in LT, moderate O_3 in MT, extremely low O_3 in UT	Station-supplied SST & procedures
187, 188, 190, 191	Biomass burning	3	Enhanced O_3 in LT, high O_3 in MT, low O_3 in UT	JOSIE-supplied SST & WMO procedures
189	Maritime background	2	Low ozone in LT, enhanced ozone in MT and extreme low ozone in UT	JOSIE-supplied SST & WMO procedures

* Due to a problem with the ESC, Simulation 176 only recorded profiles to 15 km.

** In Figure 4, 1 = blue, 2= green, 3= red

Table 5: Total and partial column statistics from two SHADOZ simulations and means for all 10 simulations (five each in Sessions 1 and 2). All simulations use SHADOZ SOPs.

Instrument	Sim 171 (DU)	Sim 182 (DU)	Mean OPM/Sonde Ratio: TCO	Mean OPM/Sonde Ratio: Trop O_3 (0-15 km)	Mean OPM/Sonde Ratio: TTL O_3 (12-18 km)	Mean OPM/Sonde Ratio: Strat O_3 (15 km-end)
OPM	282	-----	337 DU	47.0 DU	4.93 DU	298 DU
Participant 1	1.07	-----	1.03	1.09	1.02	1.04
Participant 2	1.09	-----	1.04	1.09	1.03	1.04
Participant 3	1.03	-----	1.03	1.02	0.95	1.03
Participant 4	1.01	-----	1.02	1.06	1.01	1.02
OPM	-----	334	313 DU	41.0 DU	5.30 DU	271 DU
Participant 5	-----	1.00	1.03	0.85	0.77	1.03
Participant 6	-----	1.04	1.04	0.89	0.87	1.05
Participant 7	-----	1.07	1.04	0.93	0.93	1.05
Participant 8	-----	1.00	1.02	0.88	0.87	1.02

Table 6: Total and partial column statistics from profile simulations, relative to OPM, categorized by SOP and sonde/solution types.

Methodology	No.	Mean Sonde/OPM TCO	Mean Sonde/OPM Trop O_3 (0-15 km)	Mean Sonde/OPM TTL O_3 (12-18 km)	Mean Sonde/OPM Strat O_3 (20 km-end)
SHADOZ SOP	40	1.03	1.01	0.94	1.04
JOSIE SOP	40	0.97	0.99	0.94	0.97
ENSCI 1.0%, 0.1B*	25	0.98	1.00	0.97	0.98
SPC 1.0%, 0.1B	10	0.97	0.96	0.90	0.98
ENSCI 0.5%, 0.5B	20	1.03	1.00	0.91	1.04
SPC 1.0%, 1.0B	15	1.03	1.01	0.95	1.04
ENSCI 2.0%, 0.1B	5	0.97	1.01	0.97	0.97
SPC 2.0%, 0.1B	5	0.97	0.94	0.90	0.96

* B=Buffer

Figure Caption List:

Figure 1: Map of SHADOZ stations.

Figure 2(a): Session 1 participants: (1) George Brothers (NASA/WFF); (2) Kennedy Thiong'o (Kenya Met Dept.); (3) Francisco Raimundo da Silva (INPE Natal); (4) Ernesto Corrales (Univ. Costa Rica); (5) Peter von der Gathen (Alfred Wegener Institute); (6) Herman Smit (FZ Jülich); (7) Ryan Stauffer (NASA/GSFC); (8) Gary Morris (St. Edward's Univ.); (9) Gabi Nork (FZ Jülich); (10) Anne Thompson (NASA/GSFC); (11) Bryan Johnson (NOAA ESRL); (12) Tshidi Machinini (South African Weather Service); (13) Tatsumi Nakano (Japan Met Agency); (14) Rhonie Wolff (NASA/WFF).

Figure 2(b): Session 2 participants: (1) Gonzague Romanens (MeteoSwiss); (2) Torben Blomel (FZ Jülich); (3) Jennifer Gläser (FZ Jülich); (4) Nguyen Thi Hoang Anh (Vietnam Meteorological and Hydrological Administration); (5) Anne Thompson (NASA/GSFC); (6) Jonathan Davies (Env. Climate Change Canada); (7) Zamuna Zainal (Met Malaysia); (8) Patrick Neis (FZ Jülich); (9) Gabi Nork (FZ Jülich); (10) Rigel Kivi (FMI); (11) Rene Stübi (MeteoSwiss); (12) Patrick Cullis (NOAA ESRL); (13) Herman Smit (FZ Jülich); (14) Marc Allaart (KNMI); (15) Roeland Van Malderen (Royal Meteorological Institute of Belgium); (16) Jacquelyn Witte (NASA/GSFC); (17) George Paiman (Met Dept. of Suriname); (18) Andreas Petzold (FZ Jülich); (19) Gilbert Levrat (MeteoSwiss); (20) Françoise Posny (Univ. of La Réunion).

Figure 3: (a) Schematic of an electrochemical concentration cell (ECC) in operational mode. (b) ECC instrument in Styrofoam box in which it is housed during JOSIE tests or in deployment (when launched the sensor is sealed with a Styrofoam lid). Instrument and solution type for each JOSIE-SHADOZ station appear in Tables 2 and 3, respectively.

Figure 4: Simulated ozone profiles (in partial pressure) as a function of simulation time for the troposphere and stratosphere until 33 km altitude (a) and up to 20 km in (b). Three different tropospheric ozone profiles with extreme low ozone concentrations up to the tropopause (Altitude ≈ 18 km) in blue and two profiles with moderate to enhanced middle tropospheric ozone values in green and red, respectively.

Figure 5: Ozone "raw" profiles of typical simulations in Sessions 1 (a) and 2 (b). Participants are listed in Table 3, simulation specifications are listed in Table 4.

Figure 6: (a) Participant mean profiles relative to OPM in partial pressure (mPa), and (b) % deviation (Sonde – OPM / OPM). Based on 5 simulations per participant.

Figure 7: Same as Fig. 6, except for JOSIE SOP as described in Table 4.

Figure 8: (a) Session 1 SHADOZ SOP (blue) and JOSIE SOP (red) mean profiles subtracted from the OPM profile mean; (b) Session 2 JOSIE 2.0.1 (black) and JOSIE 1.0.1 (red) SST profile means subtracted from the OPM; and (c) Session 1 and 2 mean profiles of

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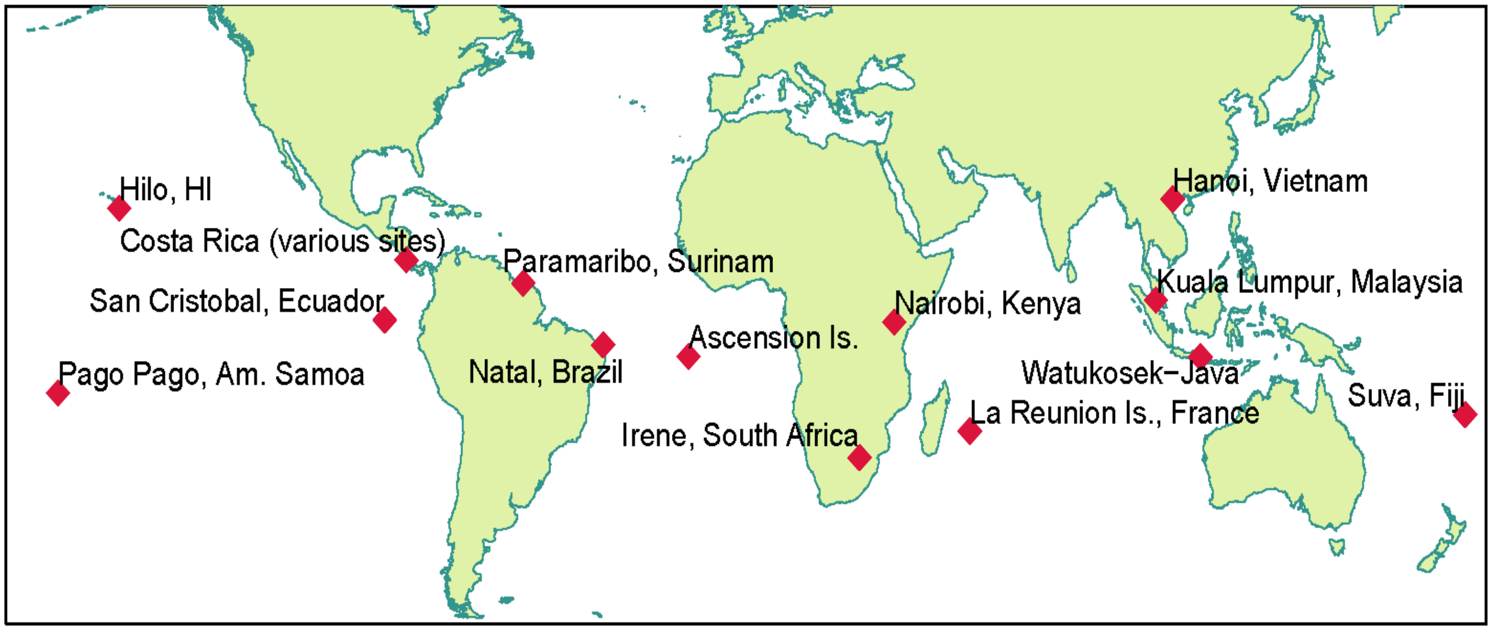
Figure SB1. Ozone and temperature profiles from a typical SHADOZ sounding at Natal, Brazil, taken from the archive, https://tropo.gsfc.nasa.gov/shadoz.

Figure SB2: (a) Set up for the simulation of vertical ozone soundings with a schematic of the Environmental Simulation Chamber, showing Ozone Photometer (OPM) standard reference, control systems, placement of four ozonesondes ("TEO") in the chamber and data acquisition system (DAS). (b) Photo of the chamber and DAS computer.



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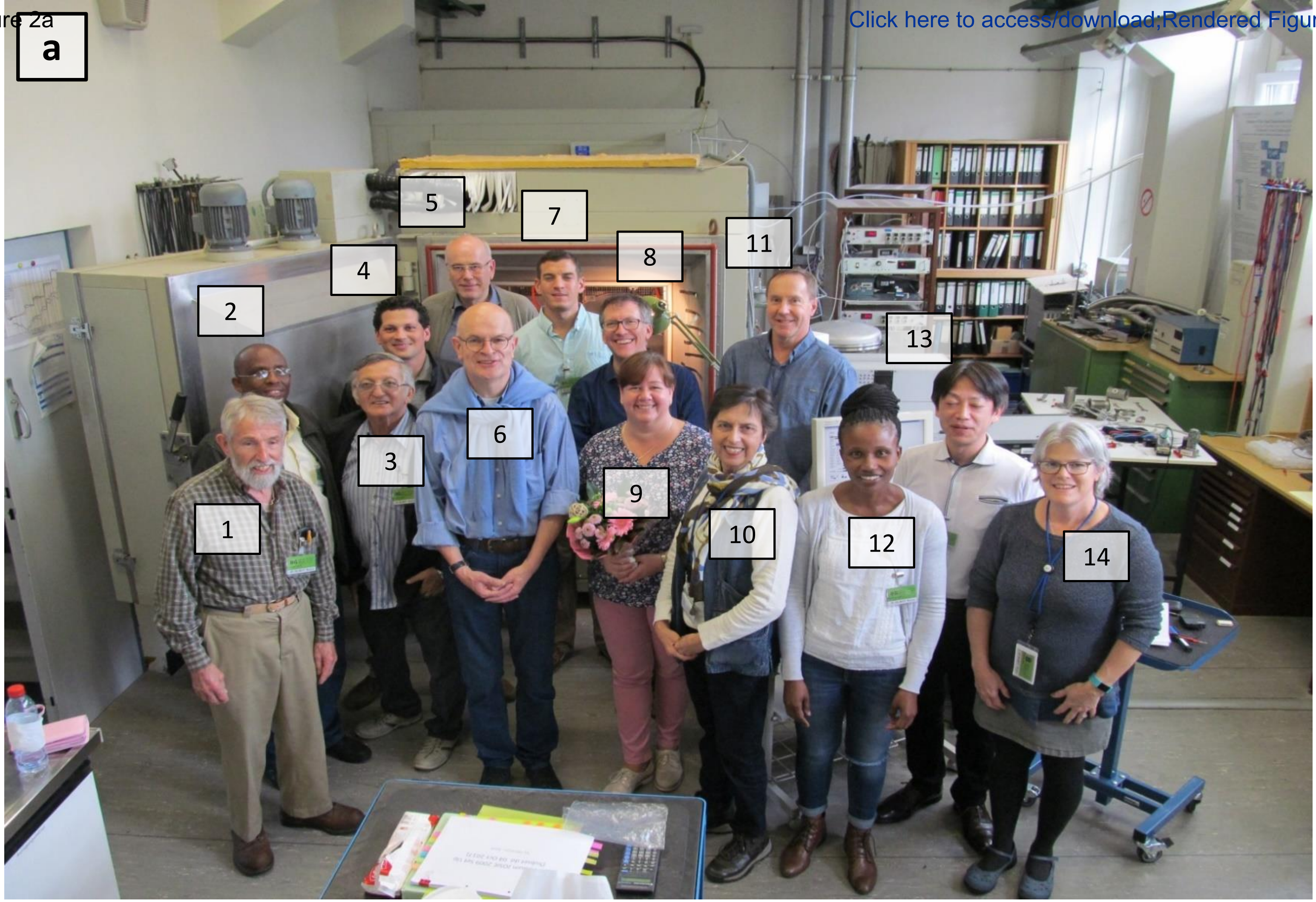

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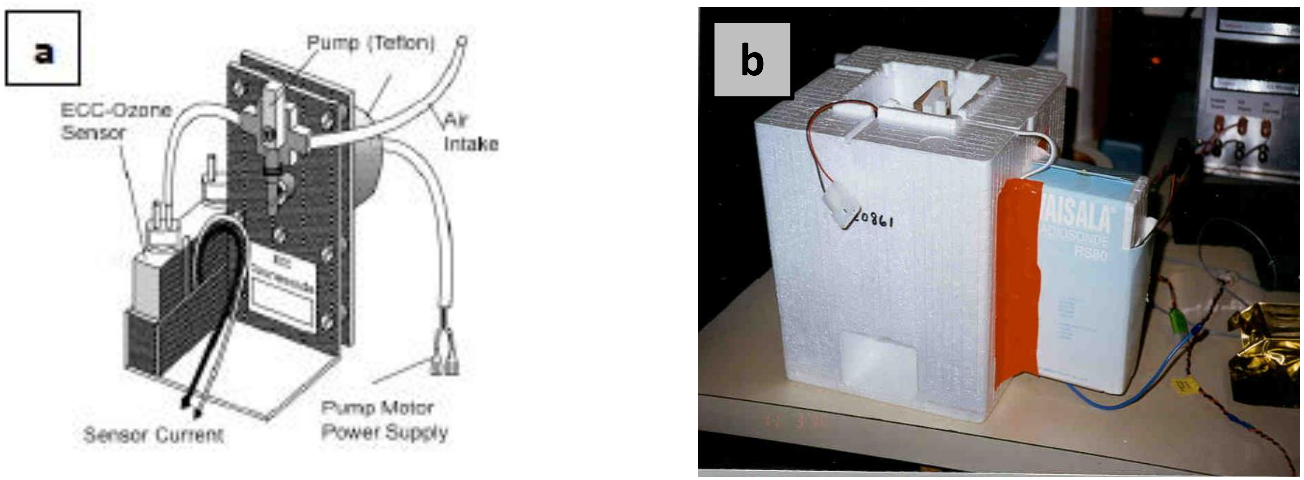

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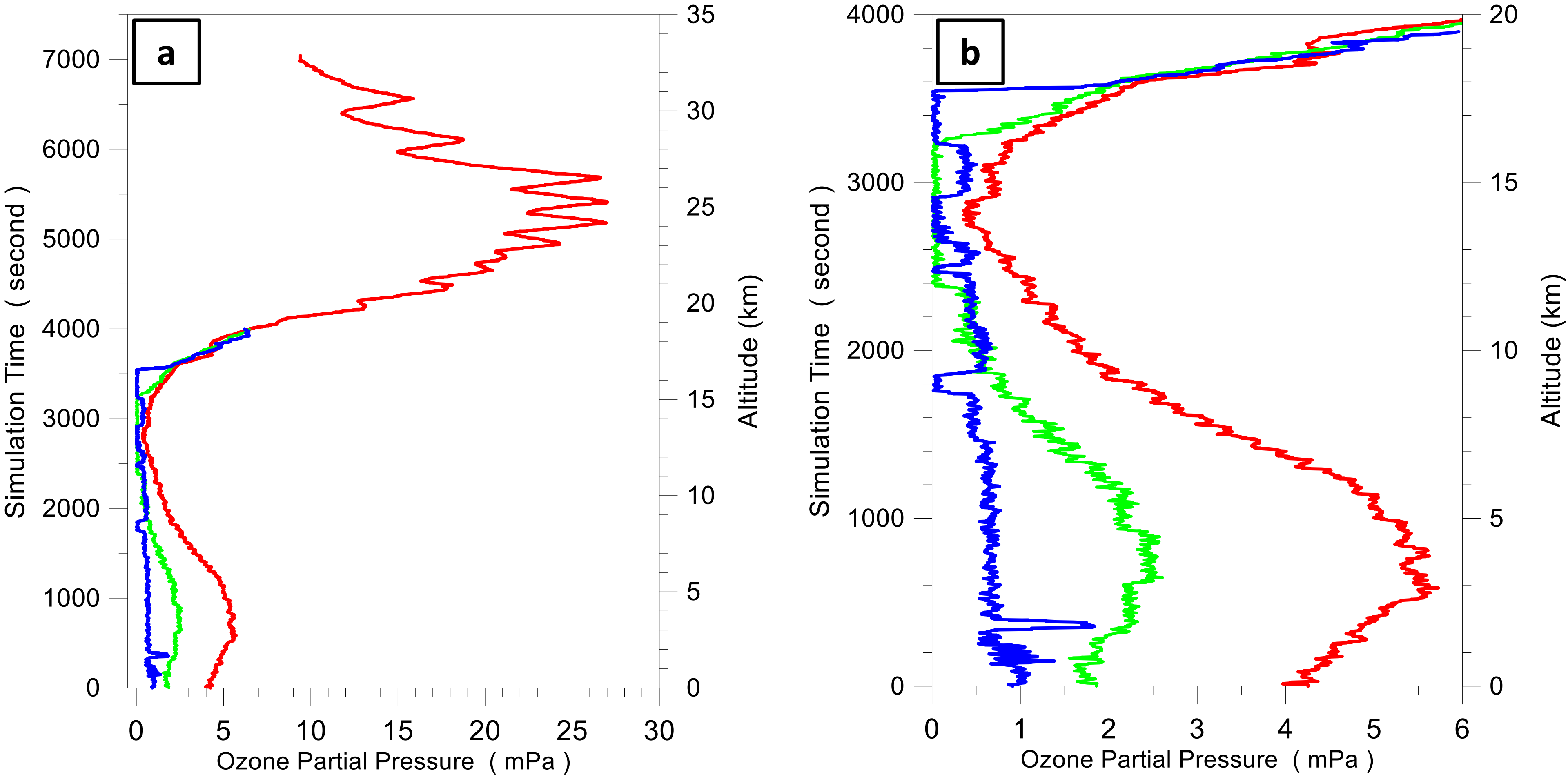

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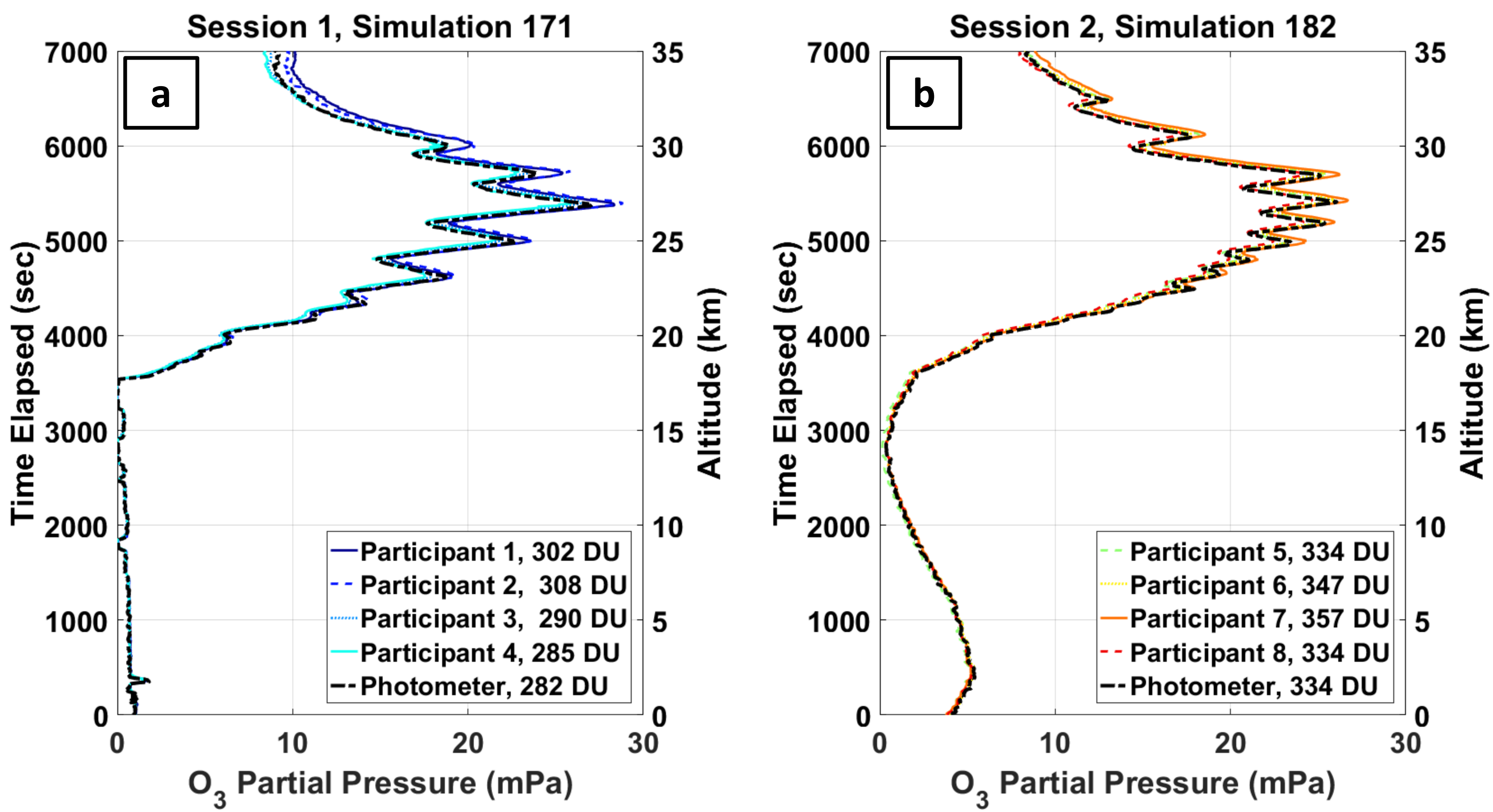

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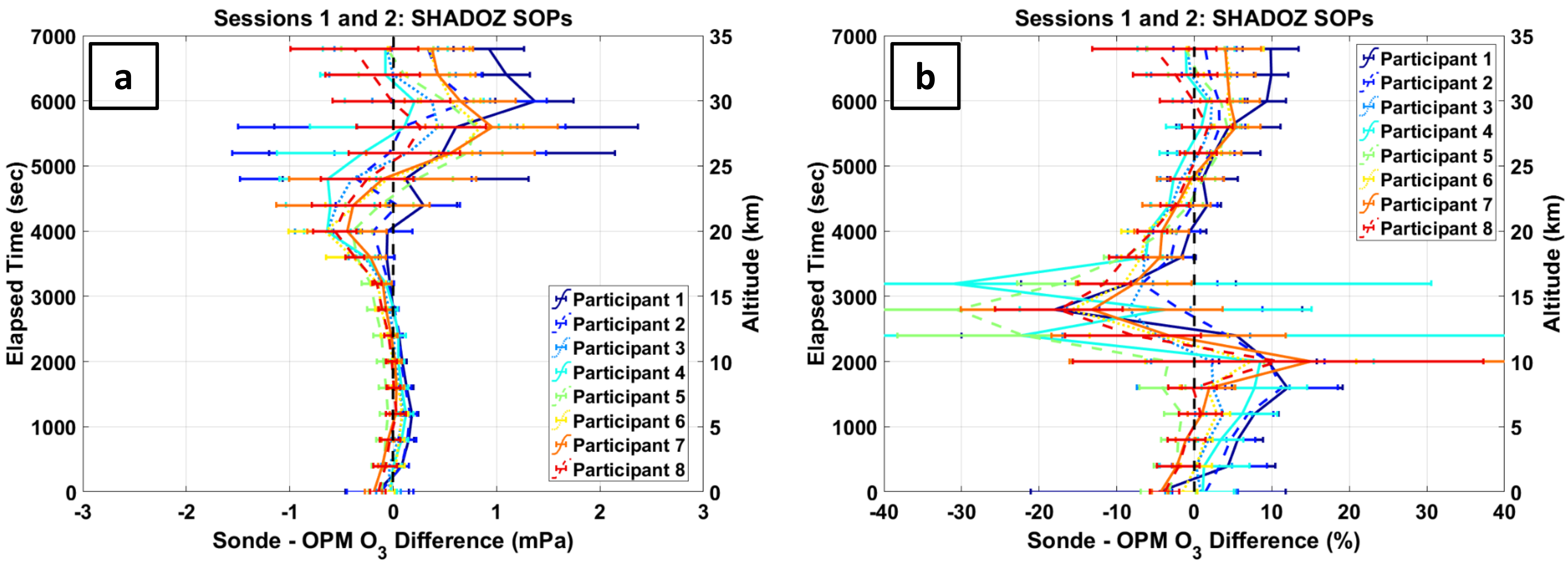

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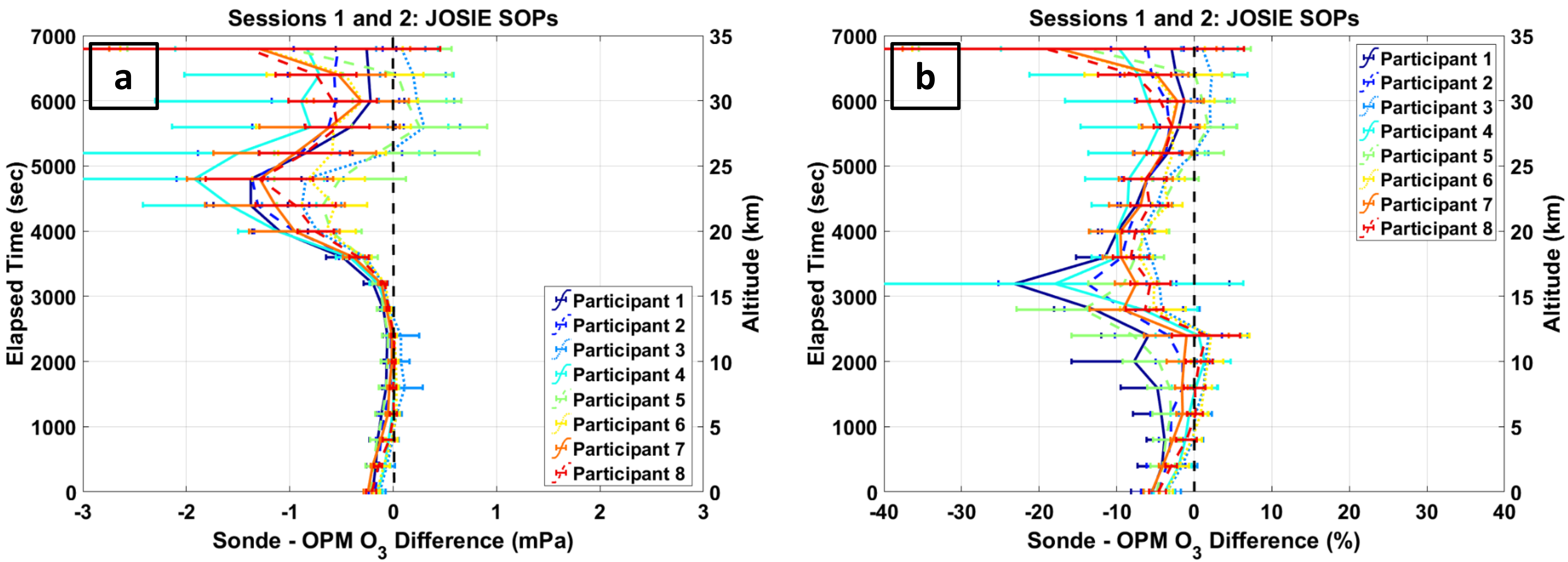

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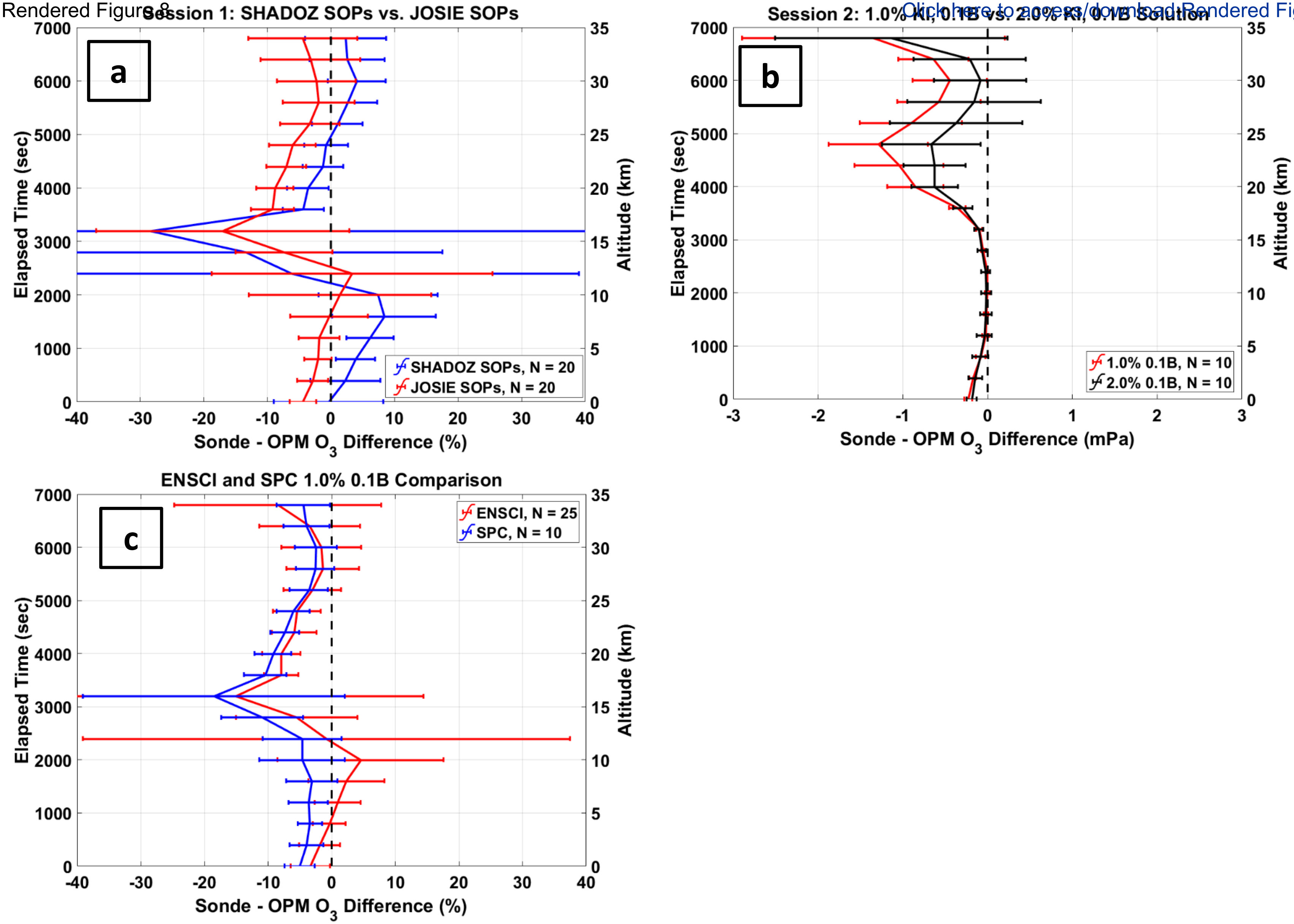

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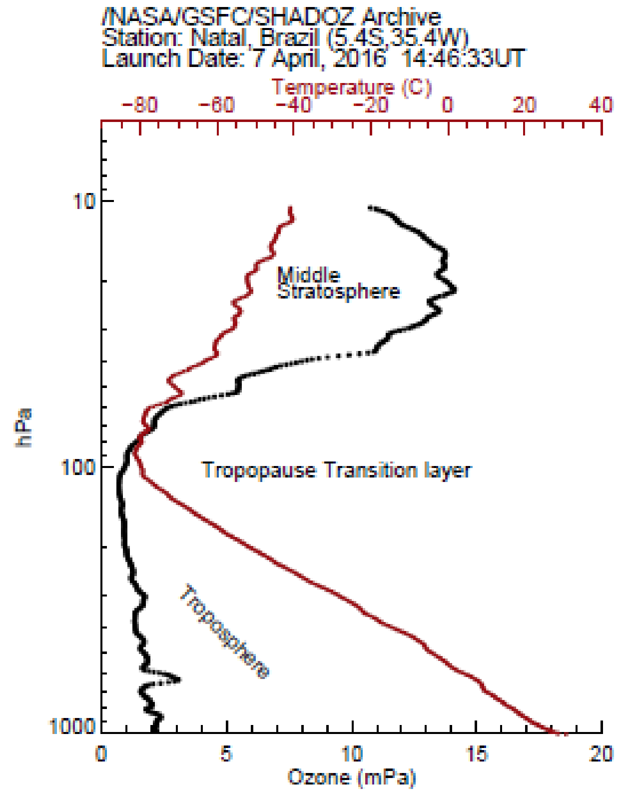

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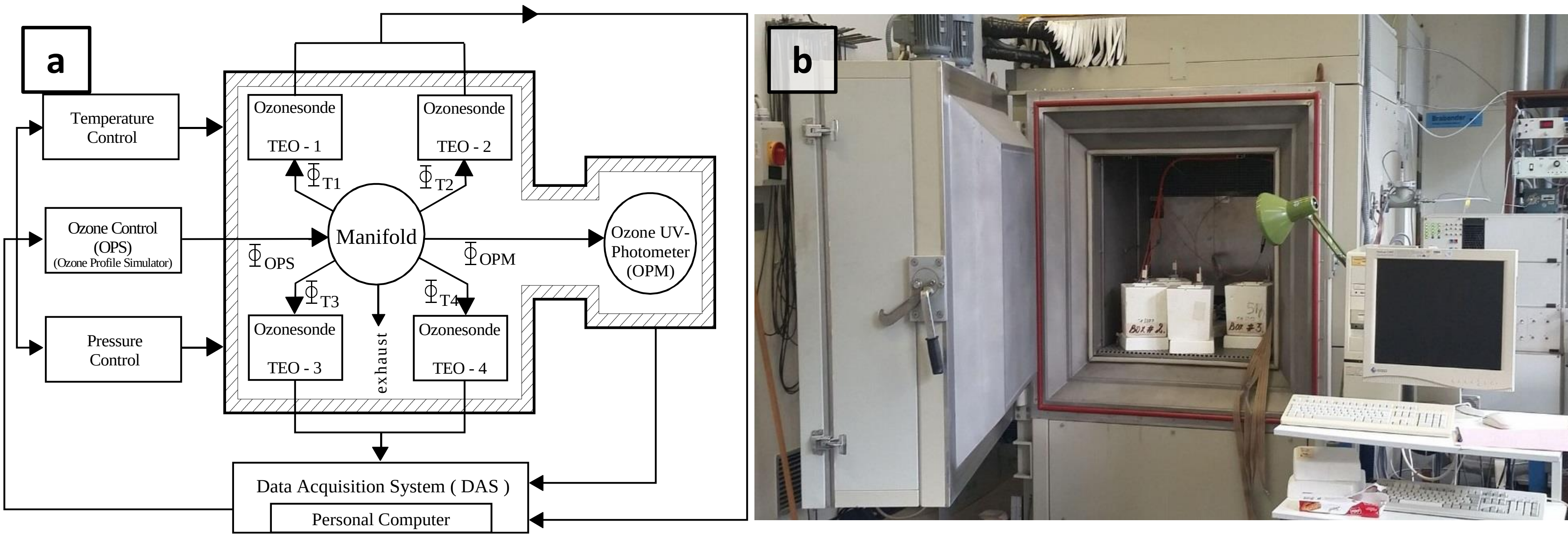

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